

# Activity induced phase separation

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We consider a mixture of passive (i.e., Brownian) and active (e.g., bacterial or colloidal swimmers) particles, and analyze the stability conditions of either uniformly mixed or phase segregated steady states consisting of phases enriched with different types of particles. We show that in sufficiently dilute mixtures the system behaves as if it were exposed to two separate heat baths of uneven temperatures. It can be described within a second virial approximation neglecting three body and higher order collisions. In this approximation, we define non-equilibrium “chemical potentials” whose gradients govern diffusion fluxes and a non-equilibrium “osmotic pressure”, which governs the mechanical stability of the interface.

**Introduction.** Suspensions of actively moving particles, performing mechanical work at the expense of internal or external energy consumption, have attracted much attention over the last years [1–9]. The interest is motivated by biological applications, but these studies shed also light on the fundamentals of statistical mechanics. These systems share many interesting properties such as spontaneous flows [10], but one of the most exciting phenomena is phase segregation [1, 2, 7–9]. It is often driven by variants of the so-called quorum sensing, which is the feedback mechanism reducing the activity of a given active particle in the presence of a high concentration of other active particles. Another type of phase segregation can occur in a mixture of particles with different levels of activity (active and non-active), when one phase is enriched in active and the other one in passive particles [11, 12]. This type of active phase segregation is far less understood.

A new spin on the problem comes from Ref. [13]. These authors study eukaryotic nuclei and the spatial segregation between eu- and hetero-chromatin, i.e., between actively processed and almost silent parts of the genome. Viewing the genome as a polymer, they argue that genes which are being expressed and, therefore, subject to RNA polymerization and other active processes, should be viewed as active monomers, and silent genes are passive monomers. They then hypothesize that the observed compartmentalization between the two kinds of chromatin is a phase segregation or rather a microphase segregation [14] based on activity. Their computational model appears reasonably consistent with the data, even though the simulation replaces the “activity” by a sufficiently high effective temperature imposed on the active monomers by a separate heat bath. One striking observation is that the effective temperature must be significantly higher than the real temperature of the passive monomers (by about a factor 20).

In this work we develop a minimal analytical model of

phase segregation between active and passive particles, call them  $\mathcal{A}$  and  $\mathcal{B}$ . Our main idea is to look at systems of sufficiently low concentration, where we resort to the type of reasoning which for equilibriums systems leads to a virial expansion. We give a systematic development of the second virial approximation in which only pair collisions between particles are considered.

If the interactions are short range, at low concentration, each particle completely loses its orientational correlations in the time between collisions with other particles. We can then consider the particles with two distinct levels of activity as exposed to two different heat baths, with temperatures  $T_A \neq T_B$ . This is clearly a system far from equilibrium, with energy flowing from the hotter to the colder reservoir via the interactions between particles; more physically, energy is taken from the source of activity and dissipated into the surrounding medium via our system of particles. There are many examples of systems whose description involves two distinct temperatures, ranging from plasmas (see basics in, e.g., [15]), to spin glasses [16] and heteropolymers [17]. Other examples are given in [18–21]. In all these works (with the notable exception of Ref. [21]), the two temperatures are used to describe motions on vastly different time scales. In our system, there is no such separation of time scales, and our goal is to explore how the access to two different heat baths enhances the tendency towards phase segregation.

Our starting point is the over-damped Langevin equation

$$\zeta_i \dot{x}_i = -\partial_i U + (2T_i \zeta_i)^{1/2} \xi_i(t), \quad (1)$$

for every particle  $i$  in the system. Here  $x_i$  indicates the position of particle  $i$  (for brevity, we will make no distinction between particles in 1 or 3 dimensions). We assume that all forces acting on a particle derive from a potential energy  $U$ , while  $\partial_i$  is the derivative with respect to  $x_i$ . The friction coefficient for particle  $i$  is  $\zeta_i$ , and  $\xi_i(t)$  is

a standard zero mean and unit variance Gaussian white noise, independent for all particles  $i$ . Finally,  $T_i$  is the temperature of the heat bath interacting with particle  $i$ , it is either  $T_A$  or  $T_B$ .

A prototypical system that may be described by this model include mixtures of actively swimming bacteria with either oxygen-starved ones (which do not actively swim), or just similar sized inactive colloidal particles; another example is a mixture of passive colloidal particles with the ones capable of light-induced catalytic transformation of the solvent, like in [4]. In the jargon of the field [22, 23], these are either run-and-tumble or active Brownian particles models, sufficiently dilute to be amenable to a virial approximation. Another major assumption behind our model is that we ignore hydrodynamic interactions (Rouse model), each particle experiencing a local friction against an immobile solvent.

**Two particles.** We first examine a system with only two particles, one  $\mathcal{A}$  and one  $\mathcal{B}$ . In this case, Eq.(1) consists of two coupled Langevin equations. Dotsenko et al [21] studied this problem when the potential energy  $U(x_A, x_B)$  is a positive-definite quadratic form of its two variables. We only assume here that  $U$  depends on the distance between particles,  $r = x_A - x_B$ , i.e.,  $U = u^{AB}(r)$  and consider mostly cases where  $u^{AB}(r)$  vanishes at large  $r$ . We derive a Langevin equation for the variable  $r$ , by combining the two equations (1):

$$\zeta_r \dot{r} = F(r) + (2\zeta_r \bar{T})^{1/2} \xi_r, \quad (2)$$

where the relevant friction is  $\zeta_r = \zeta_A \zeta_B / (\zeta_A + \zeta_B)$  and the relevant temperature is the mobility-weighted average  $\bar{T} = (\zeta_B T_A + \zeta_A T_B) / (\zeta_A + \zeta_B)$ . The definition of the effective temperature  $\bar{T}$  is dictated by the condition that the noise  $\xi_r(t)$  is a zero mean, unit variance Gaussian white noise. Since  $\zeta_{A,B} > 0$  are positive,  $\bar{T}$  is always between  $T_A$  and  $T_B$ . It follows from the Langevin equation (2) that the relative distance between particles,  $r$ , in a steady state is Boltzmann distributed with the average temperature  $\bar{T}$ , despite the fact that the system remains, of course, out of equilibrium:

$$P(r) = \exp(-U(r)/\bar{T}) z^{-1}, \quad (3)$$

$z$  is here the “partition sum” ensuring normalization [27].

**Fokker-Planck equation, currents, and violation of detailed balance.** The Langevin equations (1) can be recast as a Fokker-Planck equation for the joint probability distribution of the coordinates of all particles  $P(\{x\})$  and the corresponding currents  $J_i$ :

$$\dot{P} = -\partial_i J_i, \quad J_i = -\partial_i U P / \zeta_i - T_i \partial_i P / \zeta_i, \quad (4)$$

(using the Einstein convention for the summation over repeated indices). At steady state, for two particles the probability  $P(r)$  depends only on the distance  $r = x_A - x_B$ . Then  $\partial_A P = -\partial_B P$ , as well as  $\partial_A U = -\partial_B U$ .

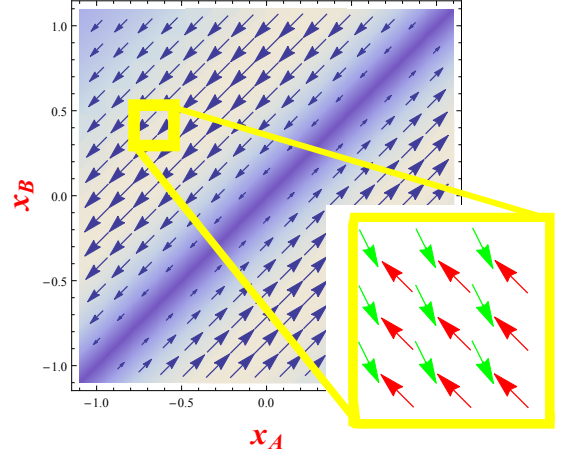


FIG. 1: Field of currents. For illustration purposes, the interaction potential is chosen in the form  $u(r) = 1/(r^2 + 1)$ , and temperatures are  $T_A = 0.5$  and  $T_B = 1$ . In the enlarged window, drift (upward and to the left) and diffusion (downward and to the right) are shown separately; unlike equilibrium system, these currents are not collinear, their vector sum gives rise to the non-potential current field.

Furthermore, under these conditions, there is no current in the  $r = x_A - x_B$  direction. The current vector  $\mathbf{J}$  must be in the perpendicular direction, which means  $J_A - J_B = 0$ , or

$$(-\partial_A U / \zeta_A + \partial_B U / \zeta_B) - (T_A \partial_A P / \zeta_A - T_B \partial_B P / \zeta_B) = 0.$$

This, of course, reproduces the Boltzmann distribution with the average temperature (3). We now compute the current  $J_A$ , taking advantage of the above result to replace the diffusion term:

$$J_A = J_B = \frac{T_A - T_B}{T_A \zeta_B + T_B \zeta_A} P(r) \partial_A U. \quad (5)$$

As expected, the current vanishes for an equilibrium system ( $T_A = T_B$ ), where detailed balance is obeyed. However, if  $T_A \neq T_B$ , detailed balance is violated. For instance, if the system is 1-dimensional, then its configuration space  $(x_A, x_B)$  is a 2-dimensional plane, and it is easy to visualize loops of current  $\mathbf{J}$  in such a plane as shown on Fig. 1. Physically, as Eq (5) suggests, these loops of current mean that the more passive particle moves, on average, mainly in the direction of the force acting on them, while the more active particle moves in the direction opposite to the force acting on them.

**Power transfer.** As we pointed out above, energy is transferred everywhere in the system from the “hot heat bath” to the “cold” bath, or from the energy source of active motion to the surrounding passive medium. The transferred power from  $\mathcal{B}$  to  $\mathcal{A}$  is the average  $w = \langle -\dot{x}_A \partial_A U \rangle$ . We note that velocity  $\dot{x}_A = J_A / P$ , while the average involves an integration with a weight

$P$ . Therefore,  $w = -\int J_A \partial_A U dx_A dx_B$ , leading to:

$$w_{B \rightarrow A} = \frac{T_B - T_A}{T_A \zeta_B + T_B \zeta_A} \int (\partial_r u^{AB}(r))^2 \frac{e^{-u^{AB}(r)/\bar{T}}}{z} dr. \quad (6)$$

As expected, this power transfer from  $B$  to  $A$  vanishes for the equilibrium system if  $T_B = T_A$ , but it is positive if  $T_B > T_A$  and negative otherwise.

To understand the meaning of the result (6), consider a 3-dimensional system (when the integration over  $dr$  runs over the volume) and an interaction potential  $U$  that does not bind the particles together. An example is a repulsive  $U(r)$  with a little bump of energy scale  $U_0$  and spatial scale  $\ell$ . Then the integral in Eq.(6) is estimated as  $(U_0/\ell)^2 \ell^3/L^3$ , with  $L^3$  the box volume (which enters in  $z$ ), and then (assuming for simplicity  $\zeta_A = \zeta_B = \zeta$ ) the result can be re-arranged as

$$w \sim [\ell (\bar{T}/\zeta) / L^3] [(T_B - T_A) U_0^2 / \bar{T}^2]. \quad (7)$$

The first factor in the square brackets is the inverse Smoluchowski time between collisions of two particles, and, therefore, the second factor is an estimate of the energy transferred during one collision.

**Many particles.** Consider now a system of  $N_i$  particles  $i$  ( $i, j = A, B$ ). The Fokker-Planck equation (4) is generalized for any number of particles. Integrating the Fokker-Planck equation over all coordinates except for one, we derive a diffusion equation for the single particle probability (proportional to the concentration) for every particle species:

$$\begin{aligned} \frac{\partial p_1^A(\mathbf{r})}{\partial t} &= \frac{N_A}{\zeta_A} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AA}}{\partial \mathbf{r}} p_2^{AA}(\mathbf{r}, \mathbf{r}') d\mathbf{r}' \right] + \\ &+ \frac{N_B}{\zeta_B} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AB}}{\partial \mathbf{r}} p_2^{AB}(\mathbf{r}, \mathbf{r}') d\mathbf{r}' \right] + \frac{T_A}{\zeta_A} \nabla_{\mathbf{r}}^2 p_1^A(\mathbf{r}). \end{aligned} \quad (8)$$

A similar equation is obtained for  $p_1^B(\mathbf{r})$ . These equations for single particle probabilities include pair probabilities  $p_2$ . By integrating the multiparticle Fokker-Planck equation over all coordinates except for two, we derive equations for  $p_2$ , which include the 3-body correlations  $p_3$ , and then a hierarchy of equations [28]. However, if the density is small enough, we can neglect the 3-body correlations and ignore all terms involving  $p_3$ , thus obtaining closed equation for  $p_2$ . Consistent with the two particle system, we obtain in this approximation:

$$p_2^{ij}(\mathbf{r}, \mathbf{r}') = p_1^i(\mathbf{r}) p_1^j(\mathbf{r}') \exp[-u^{ij}(\mathbf{r} - \mathbf{r}')/T_i] \quad (9)$$

where  $i, j = A, B$ . The effective temperatures entering these expressions is different for the three types of interactions:  $T_{AA} = T_A$ ,  $T_{BB} = T_B$  and  $T_{AB} = \bar{T}$ .

It is important to note that these distributions form only as a result of an averaging over many collisions happening in the system under steady state conditions (similar in this respect to an equilibrium system).

Inserting the *ansatz* (34) into equations (29) and introducing the concentrations  $c^i(\mathbf{r}) = N_i p_1^i(\mathbf{r})$ , we obtain closed equations for the concentrations:

$$\frac{\partial c^i(\mathbf{r})}{\partial t} = \frac{1}{\zeta_i} \frac{\partial}{\partial \mathbf{r}} \left( c^i \frac{\partial \mu_i}{\partial \mathbf{r}} \right) \quad (10)$$

These equations look like regular diffusion equations, but they are governed by *non-equilibrium* analogs of chemical potentials [29]:

$$\mu_A = T_A \ln c^A + T_A B_A c^A + \bar{T} B_{AB} c^B \quad (11)$$

and a similar equation for  $\mu_B$ . The virial coefficients are defined each with its own temperature, as  $B_{ij} = \int [1 - e^{-u^{ij}(\mathbf{r})/T_{ij}}] d^3\mathbf{r}$ .

Non-equilibrium chemical potentials, as quantities whose gradient determines the flux, were discussed in Ref. [5]. It was shown, that, unlike its equilibrium counterpart, a non-equilibrium chemical potential, in general, cannot be obtained as a derivative of a free energy. This was shown in particular for the gradient terms. In our case, the situation is different, because the non-equilibrium chemical potentials  $\mu_A$  and  $\mu_B$  appear to be the partial derivatives  $\mu_i = \frac{\partial f}{\partial c^i}$  of a function, which looks like a two-temperature free energy (per unit volume):

$$\begin{aligned} f &= T_A c^A \ln(c^A/e) + T_B c^B \ln(c^B/e) + \\ &+ (1/2) T_A B_A c_A^2 + (1/2) T_B B_B c_B^2 + \bar{T} B_{AB} c^A c^B. \end{aligned} \quad (12)$$

#### Instability of the uniform state and “spinodal.”

Suppose that  $c_0^A$  and  $c_0^B$  are the averaged spatially uniform concentrations of both components. By introducing small space dependent perturbations  $c^i(\mathbf{r}) = c_0^i + \delta c^i(\mathbf{r})$ , we perform a linear stability analysis in the standard way. This shows that an instability occurs macroscopically under the condition

$$\frac{\phi_A}{1 + \phi_A} \frac{\phi_B}{1 + \phi_B} > \frac{T_A T_B}{\bar{T}} \frac{B_{AB}^2}{B_A B_B}, \quad (13)$$

where we have defined the volume fractions  $c^i B_i = \phi_i$ . In general, the virial coefficients depend on temperature in a complex way. A simple limit corresponds to purely excluded volume interaction potentials such that the  $B$ 's do not depend on temperature. We study this case in the following.

In the plane  $\phi_A$  and  $\phi_B$ , the non-equilibrium equivalent of the spinodal line (13) is a hyperbola (see figure 2). The contrast between temperatures favors instability, it works in the same direction as contrast between interactions. But this instability, to have a physical meaning, must occur at  $\phi_A < 1$  and  $\phi_B < 1$ ; moreover,  $\phi_A + \phi_B < 1$ . For instance, consider the most symmetric case of identical particles in all respects except driven by different temperatures:  $\zeta_A = \zeta_B$ ,  $B_A = B_B = B_{AB}$ . In this case the spinodal line is in the physical range  $\phi_A + \phi_B < 1$

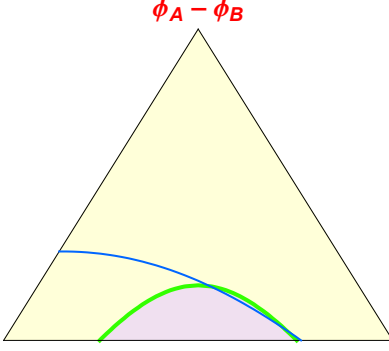


FIG. 2: Phase diagram,  $\phi_A$  and  $\phi_B$  are volume fractions of  $\mathcal{A}$ - and  $\mathcal{B}$ -particles, while  $1 - \phi_A - \phi_B$  (which is the distance to lower side of the triangle) is the fraction of solvent. Green line is the “spinodal” (below this line, uniformly mixed state is unstable). Blue line corresponds to constant osmotic pressure, it shows the possibility of two states coexisting.

if the ratio of the two temperatures is outside the range  $17 - 12\sqrt{2} \approx 0.029 < \frac{T_B}{T_A} < 17 + 12\sqrt{2} \approx 34$  [30]. Thus, a numerically large temperature contrast is required to achieve an instability by temperature difference alone. This is somewhat consistent with numerical observation of Ganai et al [13], as they used  $T_B/T_A = 20$ .

**Pressure and “binodal.”** To address not only the loss of stability of the uniform mixed state, but also the steady state phase segregation, in addition to non-equilibrium chemical potentials we also need a non-equilibrium equivalent of the osmotic pressure. Given that non-equilibrium chemical potentials are the derivatives of a “quasi-free-energy” (12), we can expect the osmotic pressure to be given by Gibbs-Duhem formula  $p = c^A \mu_A + c^B \mu_B - f$ , yielding

$$p = T_A c^A + T_B c^B + (1/2) T_A B_A c_A^2 + (1/2) T_B B_B c_B^2 + \bar{T} B_{AB} c^A c^B. \quad (14)$$

We have directly derived this result in two independent ways, first by computing the force exerted on the wall by replacing the wall with a potential ramp, not necessarily the same for both particle species,  $u_1^A(\mathbf{r})$  and  $u_1^B(\mathbf{r})$ , and second by relating the pressure to the pair correlation function in the bulk [24, 25]. Since we do not consider non-spherical particles which experience a torque upon interactions with wall, we do not have the complications studied in the recent works [22, 23, 26], and, indeed, the two derivations [31] yield identical results for the osmotic pressure (14). This result means that the densities of two coexisting phases at steady state are found by the Maxwell common tangent construction based upon the quasi-free energy function (12). So far, we make this statement in the second virial approximation only. It remains to be seen whether this is still true or not when higher order collisions are taken into account, which is necessary at higher densities.

The calculation of the dissipation (6), can be generalized to the dissipation per unit volume of the solution. The result is

$$w = c^A c^B \frac{T_B - T_A}{T_A \zeta_B + T_B \zeta_A} \int \left( \frac{\partial u^{AB}}{\partial \mathbf{r}} \right)^2 e^{-\frac{u^{AB}(\mathbf{r})}{T}} d^3 \mathbf{r}. \quad (15)$$

It shows that in a phase separated system the dissipation mostly happens around the phase boundary [32].

**Conclusion and discussion.** To conclude, we first have to estimate the temperatures  $T_A$  and  $T_B$  in terms of real parameters of active particles. Since we assumed that the re-orientation time of one particle is much smaller than the time between collisions:  $\tau_r \ll \tau_c$ , the particle trajectory between collisions is that of a random walk, characterized by an effective diffusion constant  $D_{\text{eff}} = T_{\text{eff}}/\zeta \simeq v_0^2 \tau_r/6 + D$ , where  $v_0$  is the swimming speed,  $D = T/\zeta$  is the passive diffusion constant,  $T = T_B$  is the real ambient temperature, and  $T_{\text{eff}} = T_A$  corresponds in our theory to the temperature of the hotter heat bath. The temperature difference, which controls dissipation rate (15), is therefore given by  $T_A - T_B \simeq v_0^2 \tau_r \zeta/6$ , it is directly related to the level of activity measured by the swimming speed  $v_0$ .

Returning to the applicability condition  $\tau_r \ll \tau_c$ , for particles of size  $b$  with typical distance between particles  $d$ , the collision time is estimated by the Smoluchowski formula  $\tau_c \sim D_{\text{eff}} b/d^3 \sim D_{\text{eff}} \phi/b^2$ , where  $\phi \sim b^3/d^3$  is the volume fraction of particles. The condition of applicability is then conveniently formulated in terms of the Peclet number  $v_0 \tau_r/b \equiv \text{Pe} \ll 1/\sqrt{\phi}$ . Thus, our theory should work if the system is dilute enough and/or the active drive is not too strong.

Another significant limitation of our approach is the fact that we neglect hydrodynamic interactions. This might be particularly important in the case of actively swimming colloids, as they usually drive themselves by creating and maintaining a train of diffusing chemicals, and accordingly their interaction upon approach and collision is hardly describable in terms of a conservative force potential, as we did here.

Despite all limitations, we believe that the theory developed here is useful because it is physically transparent and may be instructive as a source of physical intuition for these highly unusual driven systems.

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- [27] See also Supplementary Material at URL ??? regarding enhancement of joint diffusion of two particles when  $T_A \neq T_B$ .
- [28] See details about this hierarchy in Supplementary Material at URL ???
- [29] See detailed derivation of non-equilibrium chemical potentials in Supplementary Material at URL ???
- [30] See more general discussion of instability condition driven by contrasts of temperatures, frictions, and interactions in the Supplementary Material at URL ???
- [31] Details of both derivations are presented in Supplementary Material at URL ???
- [32] Interestingly, more detailed derivation of the dissipated power shows that, in the system with pairwise interactions only, the dissipated power is exactly expressed using two- and three-body correlations only, as shown in Supplementary Material at URL ???

## Activity induced phase separation: Supplementary material

In this Supplementary Material, we provide the following:

1. For two particles, we analyze their joint diffusion and show how it is enhanced by the uneven driving temperatures.
2. For many particles, we provide a detailed derivation of the hierarchy of the equations for the correlation functions and show how our second virial approximation comes out.
3. We provide detailed analysis of the instability conditions when not only temperatures are uneven, but so also frictions and interactions.
4. We provide two methods to derive the osmotic pressure.
5. We derive a more general expression for the transfer of power between the heat reservoirs that includes two- and three-body correlations.

### Two particles: “Center of friction” diffusion .

In the main text, we analyzed the relative motion of two particles  $\mathcal{A}$  and  $\mathcal{B}$  by looking at the variable  $r = x_{\mathcal{A}} - x_{\mathcal{B}}$ . It is interesting to find also how the presence of two distinct temperatures affects their joint diffusion in space. To do so, it is convenient to define their joint coordinate  $R$  in such a way that the noises in  $r$  and  $R$  are statistically independent. This is achieved by choosing

$$R = \frac{\zeta_{\mathcal{A}} T_{\mathcal{B}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} x_{\mathcal{A}} + \frac{\zeta_{\mathcal{B}} T_{\mathcal{A}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} x_{\mathcal{B}} . \quad (16)$$

Then, the Langevin equation for  $R$  reads

$$(\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}}) \dot{R} = \frac{T_{\mathcal{B}} - T_{\mathcal{A}}}{\bar{T}} F(r) + \sqrt{2(\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}}) \frac{(T_{\mathcal{A}} T_{\mathcal{B}})}{\bar{T}}} \xi_R(t) , \quad (17)$$

where  $\xi_R(t)$  is a zero mean unit variance Gaussian white noise (independent of  $\xi_r$ , as stated), and  $\bar{T} = (\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}) / (\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}})$  was defined in the main text. In an equilibrium system, at  $T_{\mathcal{A}} = T_{\mathcal{B}}$ , the inter-particle force  $F(r)$  does not couple to the joint motion. Not so out of equilibrium: since on average the force  $F(r)$  vanishes (by symmetry),  $\langle F(r) \rangle = 0$ , the  $F(r)$  term provides an additional noise driving the diffusion of the variable  $R$ . Of course, it is not a white noise, so that the dynamics of  $R$  is not a simple diffusion on time scales shorter or comparable to the correlation time of  $r$ . But on longer time scales  $R$  undergoes simple diffusion, with a diffusion coefficient which can be directly read out of the Langevin equation (17), because  $F(r(t))$  is statistically independent from  $\xi_R(t)$ :

$$D_R = \frac{T_{\mathcal{A}} T_{\mathcal{B}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} + \frac{1}{2} \left( \frac{T_{\mathcal{A}} - T_{\mathcal{B}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} \right)^2 (F^2)_{\omega=0} ; \quad (18)$$

here the power the spectrum of the force  $F(r)$  at zero frequency is

$$(F^2)_{\omega=0} = \int_{-\infty}^{\infty} \langle F(r(t)) F(r(t+\tau)) \rangle d\tau . \quad (19)$$

in order to find the power spectrum of the force, we Fourier transform the Langevin equation for  $r$  (Eq.(2) of the main text):

$$F_{\omega} = i\omega \zeta_r r_{\omega} - (2\zeta_r \bar{T})^{1/2} \xi_{\omega} , \quad (20)$$

Then multiplying it by the complex conjugate and assuming  $[\omega F_{\omega}]_{\omega=0} = 0$ , we obtain  $(F^2)_{\omega=0} = 2\zeta_r \bar{T}$ . This yields

$$D_R = \frac{T_{\mathcal{A}} T_{\mathcal{B}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} + \frac{1}{2} \frac{(T_{\mathcal{A}} - T_{\mathcal{B}})^2}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} \frac{\zeta_{\mathcal{A}} \zeta_{\mathcal{B}}}{(\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}})^2} . \quad (21)$$

We see that the difference in temperatures, independently of the sign, enhances the joint diffusion.

It is also instructive to write the Fokker-Planck equation in terms of the variables  $r$  and  $R$ :

$$\begin{aligned} \partial_t P = & - \left[ \frac{\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}}}{\zeta_{\mathcal{A}} \zeta_{\mathcal{B}}} \right] \partial_r (F(r) P) - \\ & - \left[ \frac{T_{\mathcal{B}} - T_{\mathcal{A}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} \right] F(r) \partial_R P + \\ & + \left[ \frac{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}}{\zeta_{\mathcal{A}} \zeta_{\mathcal{B}}} \right] \partial_r^2 P \\ & + \left[ \frac{T_{\mathcal{A}} T_{\mathcal{B}}}{\zeta_{\mathcal{A}} T_{\mathcal{B}} + \zeta_{\mathcal{B}} T_{\mathcal{A}}} \right] \partial_R^2 P . \end{aligned} \quad (22)$$

Here, the right hand side has explicitly the form of a divergence, and can be written as

$$\partial_t P = -\partial_r J_r - \partial_R J_R , \quad (23)$$

where the components of the flux are

$$J_r = \left[ \frac{\zeta_A + \zeta_B}{\zeta_A \zeta_B} \right] F(r) P - \left[ \frac{\zeta_A T_B + \zeta_B T_A}{\zeta_A \zeta_B} \right] \partial_r P, \quad (24a)$$

$$J_R = \left[ \frac{T_B - T_A}{\zeta_A T_B + \zeta_B T_A} \right] F(r) P - \left[ \frac{T_A T_B}{\zeta_A T_B + \zeta_B T_A} \right] \partial_R P. \quad (24b)$$

### Hierarchy of equations for the correlation functions

#### Notations, definitions and symmetries

As stated in the main text, we operate with a multi-dimensional Fokker-Planck equation for the probability density  $P$  as a function of the positions of all the particles in the system. More specifically, consider a system of  $N_A$  particles  $\mathcal{A}$  and  $N_B$  particles  $\mathcal{B}$ ; their coordinates are  $\mathbf{r}_1^A, \mathbf{r}_2^A, \dots, \mathbf{r}_{N_A}^A$  and  $\mathbf{r}_1^B, \mathbf{r}_2^B, \dots, \mathbf{r}_{N_B}^B$ . The potential energy includes single particle potentials and pairwise additive interactions:

$$\begin{aligned} U(\mathbf{r}_1^A, \mathbf{r}_2^A, \dots, \mathbf{r}_{N_A}^A; \mathbf{r}_1^B, \mathbf{r}_2^B, \dots, \mathbf{r}_{N_B}^B) &= \\ &= \sum_i^{N_A} u_1^A(\mathbf{r}_i^A) + \sum_j^{N_B} u_1^B(\mathbf{r}_j^B) + \\ &+ \frac{1}{2} \sum_{i \neq j}^{N_A} u^{AA}(\mathbf{r}_i^A - \mathbf{r}_j^A) + \\ &+ \sum_i^{N_A} \sum_j^{N_B} u^{AB}(\mathbf{r}_i^A - \mathbf{r}_j^B) + \\ &+ \frac{1}{2} \sum_{i \neq j}^{N_B} u^{BB}(\mathbf{r}_i^B - \mathbf{r}_j^B). \end{aligned} \quad (25)$$

The probability density  $P$  is also a function of all the coordinates:  $P = P(\mathbf{r}_1^A, \mathbf{r}_2^A, \dots, \mathbf{r}_{N_A}^A; \mathbf{r}_1^B, \mathbf{r}_2^B, \dots, \mathbf{r}_{N_B}^B)$ . It is normalized:

$$\int P(\mathbf{r}_1^A, \mathbf{r}_2^A, \dots, \mathbf{r}_{N_A}^A; \mathbf{r}_1^B, \mathbf{r}_2^B, \dots, \mathbf{r}_{N_B}^B) d\{\mathbf{r}\} = 1. \quad (26)$$

Define single particle probability densities, two particle probability densities, etc as:

$$p_1^A(\mathbf{r}) = \int \delta(\mathbf{r}_i^A - \mathbf{r}) P(\{\mathbf{r}\}) d\{\mathbf{r}\} \quad (27a)$$

$$p_1^B(\mathbf{r}) = \int \delta(\mathbf{r}_i^B - \mathbf{r}) P(\{\mathbf{r}\}) d\{\mathbf{r}\} \quad (27b)$$

$$p_2^{AA}(\mathbf{r}, \mathbf{r}') = \int \delta(\mathbf{r}_i^A - \mathbf{r}) \delta(\mathbf{r}_j^A - \mathbf{r}') P(\{\mathbf{r}\}) d\{\mathbf{r}\} \quad (27c)$$

$$p_2^{BB}(\mathbf{r}, \mathbf{r}') = \int \delta(\mathbf{r}_i^B - \mathbf{r}) \delta(\mathbf{r}_j^B - \mathbf{r}') P(\{\mathbf{r}\}) d\{\mathbf{r}\} \quad (27d)$$

$$p_2^{AB}(\mathbf{r}, \mathbf{r}') = \int \delta(\mathbf{r}_i^A - \mathbf{r}) \delta(\mathbf{r}_j^B - \mathbf{r}') P(\{\mathbf{r}\}) d\{\mathbf{r}\} \quad (27e)$$

There are 4 types of 3-particle densities:  $p_3^{AAA}(\mathbf{r}, \mathbf{r}', \mathbf{r}'')$ ,  $p_3^{AAB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'')$ ,  $p_3^{ABB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'')$ ,  $p_3^{BBB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'')$ . All these densities are independent of  $i$  and  $j$  etc, i.e., the probability density is the same for every particle of a given species.

There are several normalization and symmetry properties ( $\mathcal{X}, \mathcal{Y} = \mathcal{A}, \mathcal{B}$ ):

$$\int p_1^{\mathcal{X}}(\mathbf{r}) d\mathbf{r} = 1, \quad (28a)$$

$$p_2^{\mathcal{X}\mathcal{X}}(\mathbf{r}, \mathbf{r}') = p_2^{\mathcal{X}\mathcal{X}}(\mathbf{r}', \mathbf{r}) \quad (28b)$$

$$p_2^{\mathcal{X}\mathcal{Y}}(\mathbf{r}, \mathbf{r}') = p_2^{\mathcal{Y}\mathcal{X}}(\mathbf{r}', \mathbf{r}), \quad (28c)$$

$$\int p_2^{\mathcal{X}\mathcal{Y}}(\mathbf{r}, \mathbf{r}') d\mathbf{r} d\mathbf{r}' = 1, \quad (28d)$$

$$\int p_2^{\mathcal{X}\mathcal{Y}}(\mathbf{r}, \mathbf{r}') d\mathbf{r}' = p_1^{\mathcal{X}}(\mathbf{r}), \quad (28e)$$

$$\int p_2^{\mathcal{X}\mathcal{Y}}(\mathbf{r}, \mathbf{r}') d\mathbf{r} = p_1^{\mathcal{Y}}(\mathbf{r}') \quad (28f)$$

#### Fokker-Planck equations for the densities

Integrating out all variables except for one, or except for two, etc, we obtain the following dynamic equations for the densities:

$$\begin{aligned} \frac{\partial p_1^A(\mathbf{r})}{\partial t} &= \frac{T_A}{\zeta_A} \nabla_{\mathbf{r}}^2 p_1^A(\mathbf{r}) + \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u_1^A(\mathbf{r})}{\partial \mathbf{r}} p_1^A(\mathbf{r}) \right] + \\ &+ \frac{N_A - 1}{\zeta_A} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_2^{AA}(\mathbf{r}, \mathbf{r}') d\mathbf{r}' \right] + \\ &+ \frac{N_B}{\zeta_B} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_2^{AB}(\mathbf{r}, \mathbf{r}') d\mathbf{r}' \right]. \end{aligned} \quad (29)$$

A similar equation is obtained for  $p_1^B(\mathbf{r})$ , which is not given for brevity. For large numbers of particles, we can

replace  $N_A - 1 \simeq N_A$ .

$$\begin{aligned}
\frac{\partial p_2^{AA}(\mathbf{r}, \mathbf{r}')}{\partial t} &= \frac{T_A}{\zeta_A} (\nabla_{\mathbf{r}}^2 + \nabla_{\mathbf{r}'}^2) p_2^{AA}(\mathbf{r}, \mathbf{r}') + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u_1^A(\mathbf{r})}{\partial \mathbf{r}} p_2^{AA}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_2^{AA}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{N_A}{\zeta_A} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}'')}{\partial \mathbf{r}} p_3^{AAA}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right] + \\
&+ \frac{N_B}{\zeta_A} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}'')}{\partial \mathbf{r}} p_3^{AAB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right] + \quad (30) \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}'} \left[ \frac{\partial u_1^A(\mathbf{r}')}{\partial \mathbf{r}'} p_2^{AA}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}'} \left[ \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}'} p_2^{AA}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{N_A}{\zeta_A} \partial_{\mathbf{r}'} \left[ \int \frac{\partial u^{AA}(\mathbf{r}', \mathbf{r}'')}{\partial \mathbf{r}'} p_3^{AAA}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right] + \\
&+ \frac{N_B}{\zeta_A} \partial_{\mathbf{r}'} \left[ \int \frac{\partial u^{AB}(\mathbf{r}', \mathbf{r}'')}{\partial \mathbf{r}'} p_3^{AAB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right].
\end{aligned}$$

A similar equation (not given here) is obtained for  $p_2^{BB}$ . But the equation for the mixed probability deserves to be written down:

$$\begin{aligned}
\frac{\partial p_2^{AB}(\mathbf{r}, \mathbf{r}')}{\partial t} &= \left( \frac{T_A}{\zeta_A} \nabla_{\mathbf{r}}^2 + \frac{T_B}{\zeta_B} \nabla_{\mathbf{r}'}^2 \right) p_2^{AB}(\mathbf{r}, \mathbf{r}') + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u_1^A(\mathbf{r})}{\partial \mathbf{r}} p_2^{AB}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_2^{AB}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{N_A}{\zeta_A} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}'')}{\partial \mathbf{r}} p_3^{ABA}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right] + \\
&+ \frac{N_B}{\zeta_A} \partial_{\mathbf{r}} \left[ \int \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}'')}{\partial \mathbf{r}} p_3^{ABB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right] + \quad (31) \\
&+ \frac{1}{\zeta_B} \partial_{\mathbf{r}'} \left[ \frac{\partial u_1^B(\mathbf{r}')}{\partial \mathbf{r}'} p_2^{AB}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{1}{\zeta_B} \partial_{\mathbf{r}'} \left[ \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}'} p_2^{AB}(\mathbf{r}, \mathbf{r}') \right] + \\
&+ \frac{N_A}{\zeta_B} \partial_{\mathbf{r}'} \left[ \int \frac{\partial u^{BA}(\mathbf{r}', \mathbf{r}'')}{\partial \mathbf{r}'} p_3^{ABA}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right] + \\
&+ \frac{N_B}{\zeta_B} \partial_{\mathbf{r}'} \left[ \int \frac{\partial u^{BB}(\mathbf{r}', \mathbf{r}'')}{\partial \mathbf{r}'} p_3^{ABB}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d\mathbf{r}'' \right].
\end{aligned}$$

The equations for  $p_3$  involve  $p_4$ , and so on, *ad infinitum*.

If the density is small enough, we can neglect all triple collisions, i.e., directly discard all terms involving  $p_3$ . Indeed, given the normalization, any term containing  $p_2$  in equations (30) or (30) is of order  $1/V^2$ , while every term containing  $p_3$  is of order  $N/V^3$ . This, of course, simplifies the equations quite dramatically, and reduces them

essentially to what we obtained for the case of two particles. Omitting the single particle potential terms, we obtain

$$\begin{aligned}
\frac{\partial p_2^{AA}(\mathbf{r}, \mathbf{r}')}{\partial t} &= \frac{T_A}{\zeta_A} (\nabla_{\mathbf{r}}^2 + \nabla_{\mathbf{r}'}^2) p_2^{AA}(\mathbf{r}, \mathbf{r}') + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_2^{AA}(\mathbf{r}, \mathbf{r}') \right] + \quad (32) \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}'} \left[ \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}'} p_2^{AA}(\mathbf{r}, \mathbf{r}') \right],
\end{aligned}$$

(and a similar equation for  $p_2^{BB}$ ),

$$\begin{aligned}
\frac{\partial p_2^{AB}(\mathbf{r}, \mathbf{r}')}{\partial t} &= \left( \frac{T_A}{\zeta_A} \nabla_{\mathbf{r}}^2 + \frac{T_B}{\zeta_B} \nabla_{\mathbf{r}'}^2 \right) p_2^{AB}(\mathbf{r}, \mathbf{r}') + \\
&+ \frac{1}{\zeta_A} \partial_{\mathbf{r}} \left[ \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_2^{AB}(\mathbf{r}, \mathbf{r}') \right] + \quad (33) \\
&+ \frac{1}{\zeta_B} \partial_{\mathbf{r}'} \left[ \frac{\partial u^{AB}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}'} p_2^{AB}(\mathbf{r}, \mathbf{r}') \right].
\end{aligned}$$

These equations are simple enough to guess the solution based on our knowledge of the two-particle case:

$$p_2^{AA}(\mathbf{r}, \mathbf{r}') = p_1^A(\mathbf{r}) p_1^A(\mathbf{r}') \exp \left[ -\frac{u^{AA}(\mathbf{r} - \mathbf{r}')}{T_A} \right] \quad (34a)$$

$$p_2^{BB}(\mathbf{r}, \mathbf{r}') = p_1^B(\mathbf{r}) p_1^B(\mathbf{r}') \exp \left[ -\frac{u^{BB}(\mathbf{r} - \mathbf{r}')}{T_B} \right] \quad (34b)$$

$$p_2^{AB}(\mathbf{r}, \mathbf{r}') = p_1^A(\mathbf{r}) p_1^B(\mathbf{r}') \exp \left[ -\frac{u^{AB}(\mathbf{r} - \mathbf{r}')}{\bar{T}} \right] \quad (34c)$$

Of course, the central feature of this result is the appearance of the average temperature, as defined in the main text

$$\bar{T} = \frac{\zeta_A T_B + \zeta_B T_A}{\zeta_A + \zeta_B}, \quad (35)$$

In order to obtain the result (34), we look for a solution of the form  $p_2^{XY}(\mathbf{r}, \mathbf{r}') = q^{XY}(\mathbf{r}, \mathbf{r}') \exp [-\beta u^{XY}(\mathbf{r}, \mathbf{r}')]$ , plug it into equations (32) or (33), and discover, that in the remaining equation for  $q$ , the variables separate, meaning that  $q$  factorizes into a factor that depends only on  $\mathbf{r}$  and a factor that depends only on  $\mathbf{r}'$ .

### Diffusion equations and non-equilibrium chemical potentials

Plugging the *ansatz* (34) into equations (29), we obtain closed results for the densities. This involves the integral

$$\int \frac{\partial u^{AA}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} p_1^A(\mathbf{r}') e^{-\frac{u^{AA}(\mathbf{r}, \mathbf{r}')}{T_A}} d\mathbf{r}', \quad (36)$$

which can be integrated by parts. Finally, we obtain equations that look like diffusion equations for a regular



system in contact with a thermostat,

$$\frac{\partial c^A(\mathbf{r})}{\partial t} = \frac{1}{\zeta_A} \frac{\partial}{\partial \mathbf{r}} \left( c^A \frac{\partial \mu_A}{\partial \mathbf{r}} \right) \quad (37a)$$

$$\frac{\partial c^B(\mathbf{r})}{\partial t} = \frac{1}{\zeta_B} \frac{\partial}{\partial \mathbf{r}} \left( c^B \frac{\partial \mu_B}{\partial \mathbf{r}} \right), \quad (37b)$$

but these equations contain *non-equilibrium chemical potentials*, as stated in the main text (Eq.(11)).

### Linear stability analysis

Suppose that  $c_0^A$  and  $c_0^B$  are the averaged spatially uniform concentrations of both components. By introducing small space dependent perturbations  $c^A(\mathbf{r}) = c_0^A + \delta c^A(\mathbf{r})$  and  $c^B(\mathbf{r}) = c_0^B + \delta c^B(\mathbf{r})$ , we perform a linear stability analysis in the standard way:

$$\begin{cases} \frac{\partial \delta c^A}{\partial t} = \frac{1}{\zeta_A} \nabla^2 [(T_A + T_A c_0^A B_A) \delta c^A + (\bar{T} c_0^A B_{AB}) \delta c^B] \\ \frac{\partial \delta c^B}{\partial t} = \frac{1}{\zeta_B} \nabla^2 [(\bar{T} c_0^B B_{AB}) \delta c^A + (T_B + T_B c_0^B B_B) \delta c^B] \end{cases} \quad (38)$$

This shows that an instability occurs (at  $q = 0$ , i.e., macroscopically) under the condition that the determinant of this matrix vanishes, i.e., the system is unstable if

$$c_0^A c_0^B \bar{T}^2 B_{AB}^2 > T_A T_B (1 + c_0^A B_A) (1 + c_0^B B_B). \quad (39)$$

At the instability, the unstable combination (eigenvector whose eigenvalue flips sign) is

$$\frac{\delta c^A(\mathbf{r}) \zeta_A}{\sqrt{T_A B_A c_0^A (1 + B_A c_0^A)}} - \frac{\delta c^B(\mathbf{r}) \zeta_B}{\sqrt{T_B B_B c_0^B (1 + B_B c_0^B)}}. \quad (40)$$

In the plane  $(c_0^A, c_0^B)$ , the spinodal line (39) is a hyperbola (Fig. 3). A better way to represent it is to use a triangular phase diagram as given in Fig.2 of the main text.

### Non-equilibrium “spinodal” line for athermal particles

The dimensionless parameters of the system are as follows:

- Contrast of excluded volumes,

$$\beta = \frac{B_{AB}}{\sqrt{B_A B_B}}. \quad (41)$$

- Contrast of temperatures,

$$\tau = \frac{T_A - T_B}{T_A + T_B}; \quad -1 < \tau < 1. \quad (42)$$

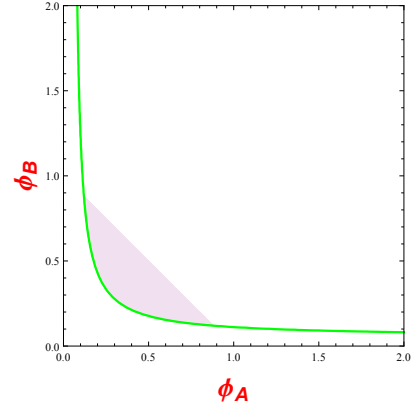


FIG. 3: Spinodal line (39) (in green), shown for one particular choice of parameters  $(T_A/T_B, \zeta_A/\zeta_B, B_A B_B/B_{AB}^2)$ . Physical meaning has only region  $\phi_A + \phi_B < 1$ , the instability region within this region is shaded.

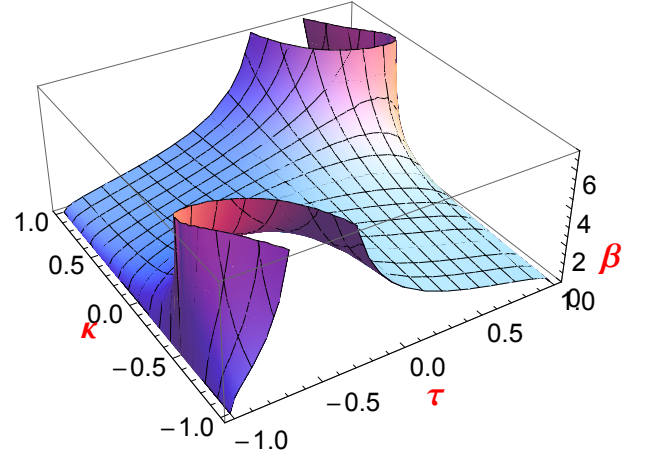


FIG. 4: Instability exists above this surface, see condition (44).

- Contrast of frictions,

$$\kappa = \frac{\zeta_A - \zeta_B}{\zeta_A + \zeta_B}; \quad -1 < \kappa < 1. \quad (43)$$

The condition that the spinodal exists within the physical range  $\phi_A + \phi_B < 1$  reads

$$\frac{4\kappa^2 \tau^2 - 4\kappa \tau + 1}{1 - \tau^2} \beta^2 > 9. \quad (44)$$

This condition is presented graphically in two different ways, in Fig. 4 in the form of a 3D surface and in Fig. 5 as an array of 2D plots. Beautifully, the contrast of frictions  $\kappa$  becomes irrelevant for the equilibrium system, when  $\tau = 0$ .

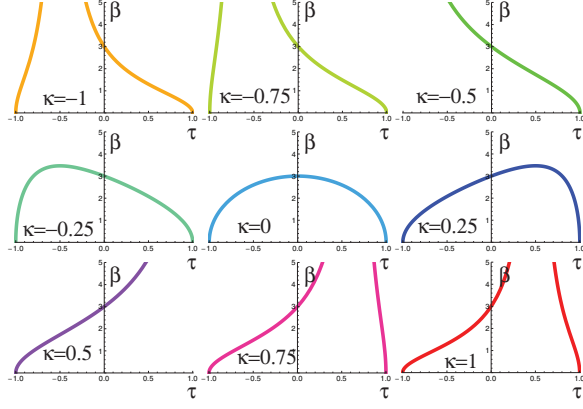


FIG. 5: For every  $\kappa$ , instability exists above the line, according to Eqns (44).

### Non-equilibrium osmotic pressure and “binodal”

To address the steady state phase segregation, in addition to the non-equilibrium chemical potentials we also need to define a non-equilibrium osmotic pressure. We derive it in two different ways.

#### Derivation 1

To find the osmotic pressure, imagine that the system “feels” single particle potentials  $u_1^A(\mathbf{r})$  and  $u_1^B(\mathbf{r})$  such that they are both like a box, except that one wall of this box has a (not necessarily very) sharp potential “ramp” in the direction, perpendicular to the wall:  $u_1^{A,B}(\mathbf{r}) = f_{A,B}x$ , as shown in Fig.6. In this case, the pressure is found according to

$$p = f_A \int_0^\infty c^A(x) dx + f_B \int_0^\infty c^B(x) dx, \quad (45)$$

because every particle  $\mathcal{A}$  present in the ramp area exerts on the wall the force  $f_A$ , and similarly for  $\mathcal{B}$ . We emphasize, that this is actually an *osmotic* pressure, in the sense that the ramp potentials  $u_1^{A,B}(\mathbf{r}) = f_{A,B}x$  act only on the  $\mathcal{A}$  and  $\mathcal{B}$  particles while the solvent penetrates everywhere completely freely. This means that our ramp potentials represent an osmotic piston.

A similar expression for the osmotic pressure was also used in Ref. [22], where it is derived from the expression of the Helmholtz partition sum, i.e., from equilibrium statistical mechanics. We feel necessary to emphasize that Eq. (45) is derived on purely mechanical grounds, and it has nothing to do with thermodynamic equilibrium. As such, it is perfectly applicable to our present problem.

To find the steady state concentration profile in the presence of ramp potentials, we slightly generalize the diffusion equations (37) by including the external poten-

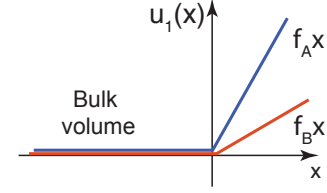


FIG. 6: Ramp potential used to calculate pressure.

tials  $u_1$ :

$$\mu_A \rightarrow \mu_A + u_1^A \text{ and } \mu_B \rightarrow \mu_B + u_1^B. \quad (46)$$

At steady state, the concentration profile must be such that  $\mu + u_1 = \text{const}$  for both the  $\mathcal{A}$  and  $\mathcal{B}$  components. This can be written as

$$c^A(\mathbf{r}) = C^A e^{-\frac{u_1^A}{T_A}} \left[ 1 + c^A(\mathbf{r}) B_A + c^B(\mathbf{r}) B_{AB} \frac{\bar{T}}{T_A} \right] \quad (47a)$$

$$c^B(\mathbf{r}) = C^B e^{-\frac{u_1^B}{T_B}} \left[ 1 + c^B(\mathbf{r}) B_B + c^A(\mathbf{r}) B_{AB} \frac{\bar{T}}{T_B} \right] \quad (47b)$$

Here  $C^A$  and  $C^B$  are normalization factors. To make things simple, we assume that the “ramps” are not too shallow, such that the normalization integral is dominated by the bulk volume  $V$  where both ramp potentials vanish. Given that the virial terms in the chemical potentials are the corrections to the ideal gas, we solve iteratively and get

$$c^A(\mathbf{r}) = \frac{N_A}{V} e^{-\frac{u_1^A}{T_A}} \left[ 1 + \frac{N_A}{V} B_A \left( 1 - e^{-\frac{u_1^A}{T_A}} \right) + \frac{N_B}{V} B_{AB} \frac{\bar{T}}{T_A} \left( 1 - e^{-\frac{u_1^B}{T_B}} \right) \right] \quad (48)$$

and similarly for  $c^B(\mathbf{r})$ . Note that the result does not depend on the ramp forces  $f_A$  and  $f_B$ , which do not have to be identical.

#### Derivation 2

Our starting point of the second derivation is the kinetic expression of the pressure

$$p = p_{\text{ideal}} - \frac{N}{6V} \left\langle \sum_{i \neq j} \mathbf{r}_{ij} \cdot \partial_{\mathbf{r}_{ij}} u(\mathbf{r}_{ij}) \right\rangle. \quad (49)$$

Sometimes it is called Irving-Kirkwood formula [24, 25]. As in the first derivation, the important point is that this equation follows from pure mechanics, does not make any assumption related to equilibrium statistical mechanics. In terms of pair distributions  $\rho_2^{ij}$  the Irving-Kirkwood

formula reads

$$\begin{aligned}
p = p_{\text{ideal}} &- \frac{N_{\mathcal{A}}^2}{6} \int \mathbf{r}_{12} \cdot \partial_{\mathbf{r}_{12}} u^{\mathcal{A}\mathcal{A}}(\mathbf{r}_{12}) p_2^{\mathcal{A}\mathcal{A}}(\mathbf{r}_1, \mathbf{r}_2) - \\
&- \frac{N_{\mathcal{A}} N_{\mathcal{B}}}{3} \int \mathbf{r}_{12} \cdot \partial_{\mathbf{r}_{12}} u^{\mathcal{A}\mathcal{B}}(\mathbf{r}_{12}) p_2^{\mathcal{A}\mathcal{B}}(\mathbf{r}_1, \mathbf{r}_2) - \\
&- \frac{N_{\mathcal{B}}^2}{6} \int \mathbf{r}_{12} \cdot \partial_{\mathbf{r}_{12}} u^{\mathcal{B}\mathcal{B}}(\mathbf{r}_{12}) p_2^{\mathcal{B}\mathcal{B}}(\mathbf{r}_1, \mathbf{r}_2) .
\end{aligned} \tag{50}$$

Using the ansatz 34 for  $p_2$ , and integrating by parts (and remembering that  $\nabla \cdot \mathbf{r} = 3$ ), we obtain the same result as before for the osmotic pressure.

### Power transfer

In the main text, we outlined the derivation of the power transfer in the cases of either two particles, or many particles with only pairwise collisions. Here we establish a more general result which suggests that the power transfer is expressed in terms of only pair and triple correlation functions (but not higher order ones). Consider the work performed by all forces per unit time on all  $\mathcal{A}$  particles, which is also the power received by  $\mathcal{A}$

particles:

$$W = \sum_i^{N_{\mathcal{A}}} \int \frac{\partial U}{\partial \mathbf{r}_i^{\mathcal{A}}} \left[ \frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}} \frac{\partial P}{\partial \mathbf{r}_i^{\mathcal{A}}} + \frac{\partial U}{\partial \mathbf{r}_i^{\mathcal{A}}} \frac{P}{\zeta_{\mathcal{A}}} \right] d\{\mathbf{r}\} , \tag{51}$$

with  $U = U(\{\mathbf{r}_j^{\mathcal{A}}\}, \{\mathbf{r}_k^{\mathcal{B}}\})$  the total potential energy of the system. In the integral (51), the first factor is the force which acts on particle  $\mathcal{A}_i$  due to all other particles, while the second factor (in square brackets) is the current, i.e., the velocity of the particle  $\mathcal{A}_i$  multiplied by the probability density  $P$ . Thus, the integral (51) is the average power transfer to one particle  $\mathcal{A}_i$ . By symmetry, it is independent of  $i$ , so that the summation over  $i$  reduces to a factor  $N_{\mathcal{A}}$ . As long as the interaction potentials are pairwise additive (25), the force is also a sum:

$$\frac{\partial U}{\partial \mathbf{r}_i^{\mathcal{A}}} = \sum_{j \neq i}^{N_{\mathcal{A}}} \frac{\partial u^{\mathcal{A}\mathcal{A}}(\mathbf{r}_i^{\mathcal{A}} - \mathbf{r}_j^{\mathcal{A}})}{\partial \mathbf{r}_i^{\mathcal{A}}} + \sum_k^{N_{\mathcal{B}}} \frac{\partial u^{\mathcal{A}\mathcal{B}}(\mathbf{r}_i^{\mathcal{A}} - \mathbf{r}_k^{\mathcal{B}})}{\partial \mathbf{r}_i^{\mathcal{A}}} . \tag{52}$$

The first term is the force acting on particle  $\mathcal{A}_i$  due to other  $\mathcal{A}$  particles, by symmetry this term vanishes on average in the sum over  $\mathcal{A}$  particles. And the second term, which is the due to  $\mathcal{B}$  particles on  $\mathcal{A}$  particles, yields:

$$\begin{aligned}
W = & \frac{N_{\mathcal{A}} T_{\mathcal{A}}}{\zeta_{\mathcal{A}}} \int \frac{\partial u^{\mathcal{A}\mathcal{B}}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \times \frac{\partial p_2^{\mathcal{A}\mathcal{B}}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} d^3 \mathbf{r} d^3 \mathbf{r}' + \frac{N_{\mathcal{A}}}{\zeta_{\mathcal{A}}} \int \left( \frac{\partial u^{\mathcal{A}\mathcal{B}}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \right)^2 p_2^{\mathcal{A}\mathcal{B}}(\mathbf{r}, \mathbf{r}') d^3 \mathbf{r} d^3 \mathbf{r}' + \\
& + \frac{N_{\mathcal{A}}^2}{\zeta_{\mathcal{A}}} \int \frac{\partial u^{\mathcal{A}\mathcal{B}}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \times \frac{\partial u^{\mathcal{A}\mathcal{A}}(\mathbf{r} - \mathbf{r}'')}{\partial \mathbf{r}} p_3^{\mathcal{A}\mathcal{B}\mathcal{A}}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d^3 \mathbf{r} d^3 \mathbf{r}' d^3 \mathbf{r}'' + \\
& + \frac{N_{\mathcal{A}} N_{\mathcal{B}}}{\zeta_{\mathcal{A}}} \int \frac{\partial u^{\mathcal{A}\mathcal{B}}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \times \frac{\partial u^{\mathcal{A}\mathcal{B}}(\mathbf{r} - \mathbf{r}'')}{\partial \mathbf{r}} p_3^{\mathcal{A}\mathcal{B}\mathcal{B}}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') d^3 \mathbf{r} d^3 \mathbf{r}' d^3 \mathbf{r}''
\end{aligned} \tag{53}$$

Neglecting the three body collisions (terms with  $p_3$ ), and using the known expression (34) for  $p_2$ , we return to the result given in the main text.

Here, we emphasize once again that, as long as interaction potentials are pairwise additive, as in Eq.(25), Eq.(53) is exact.